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(54) SILICON CARBIDE BASED POROUS COMPACT AND METHOD FOR MANUFACTURING THE SAME

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a silicon carbide based porous compact and a method for manufacturing the same which is high in porosity, high in thermal conductivity, moreover heighten in strength, as well as capable of producing with a low cost.

SOLUTION: The silicon carbide based porous compact consists of silicon carbide grain and metallic silicon to be an aggregate. A mean pore diameter of silicon carbide based porous compact is larger than a mean grain diameter of silicon carbide grains by  $\geq 2.5$  times. The silicon carbide based porous compact is manufactured by forming a material obtained by mixing and kneading a raw material of silicon carbide grain, metallic silicon and an organic binder, into predetermined shape, the obtained compact is preliminary fired to eliminate organic binder in the compact, then the compact is normally fired.

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**CLAIMS**

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[Claim(s)]

[Claim 1] The nature porous body of silicon carbide characterized by being a nature porous body of silicon carbide containing the silicon carbide particle used as the aggregate, and metal silicon, and the average pore diameter of this nature porous body of silicon carbide being 0.25 or more times of the mean particle diameter of this silicon carbide particle.

[Claim 2] The nature porous body of silicon carbide characterized by being a nature porous body of silicon carbide containing the silicon carbide particle used as the aggregate, and metal silicon, and the contact angle of this silicon carbide particle and this metal silicon being an acute angle.

[Claim 3] The nature porous body of silicon carbide which is a nature porous body of silicon carbide containing the silicon carbide particle used as the aggregate, and metal silicon, and is characterized by forming the vesicular structure when the secondary structure particle of a large number formed when these four or more silicon carbide particles contacted joins mutually together to this metal silicon of 1.

[Claim 4] The nature porous body of silicon carbide according to claim 3 whose number of this silicon carbide particle contained in this secondary structure particle of 1 is 30% or more of the number of this silicon carbide particle contained in this nature porous body of silicon carbide.

[Claim 5] The nature porous body of silicon carbide characterized by being a nature porous body of silicon carbide containing the silicon carbide particle used as the aggregate, and metal silicon, and the interfacial area of this silicon carbide particle and this metal silicon being 50% or more which doubled this interfacial area and the surface area of this metal silicon of area.

[Claim 6] The nature porous body of silicon carbide given in any 1 term of claims 1-5 which have the silicate compound phase of an amorphous substance or a crystalline substance on the outskirts of a front face or the outskirts of this silicon carbide particle and/or this metal silicon.

[Claim 7] The nature porous body of silicon carbide according to claim 6 which these silicon carbide particles have combined with this metal silicon and/or this silicate compound phase.

[Claim 8] This silicate compound phase is a nature porous body of silicon carbide according to claim 6 or 7 whose eutectic point of this metallic element more than a kind and this silicon dioxide is 1200-1600 degrees C including the metallic element and silicon dioxide more than kinds other than silicon.

[Claim 9] This silicate compound phase is a nature porous body of silicon carbide given in any 1 term of claims 6-8 whose content to the sum total of the this silicon carbide particle and this metal silicon of this metallic element more than a kind is 0.1 to 10 mass % including the metallic element and silicon dioxide more than kinds other than silicon.

[Claim 10] The nature porous body of silicon carbide according to claim 8 or 9 whose a kind is an alkaline-earth-metal element at least among these metallic elements more than a kind.

[Claim 11] The nature porous body of silicon carbide according to claim 10 which contains further metallic elements other than an alkaline-earth-metal element as this metallic element more than a kind.

[Claim 12] The nature porous body of silicon carbide according to claim 10 or 11 these whose alkaline earth metal elements are calcium and/or strontium.

[Claim 13] The nature porous body of silicon carbide given in any 1 term of claims 1-12 whose content to the sum total of the this silicon carbide particle and this metal silicon of this metal silicon is five to 50 mass %.

[Claim 14] The honeycomb structure object characterized by being constituted by any 1 term of claims 1-13 by the nature porous body of silicon carbide of a publication.

[Claim 15] The manufacture approach of the nature porous body of silicon carbide characterized by carrying out actual baking under a reduced pressure ambient atmosphere after adding metal silicon and an organic

binder in a silicon carbide particle raw material, fabricating the plastic matter mixed, and kneaded and obtained in a predetermined configuration, carrying out temporary quenching of the acquired Plastic solid and removing this organic binder in this Plastic solid.

[Claim 16] The manufacture approach of the nature porous body of silicon carbide characterized by carrying out actual baking under the reducing atmosphere containing hydrogen after adding metal silicon and an organic binder in a silicon carbide particle raw material, fabricating the plastic matter mixed, and kneaded and obtained in a predetermined configuration, carrying out temporary quenching of the acquired Plastic solid and removing this organic binder in this Plastic solid.

[Claim 17] The manufacture approach of the nature porous body of silicon carbide characterized by carrying out actual baking under a non-oxidizing atmosphere after adding the compound which contains a metallic element or this metallic element in a silicon carbide particle raw material, metal silicon, and an organic binder, fabricating the plastic matter mixed, and kneaded and obtained in a predetermined configuration, carrying out temporary quenching of the acquired Plastic solid and removing this organic binder in this Plastic solid.

[Claim 18] The manufacture approach of the nature porous body of silicon carbide according to claim 17 using this metallic element whose eutectic point with a silicon dioxide is 1200-1600 degrees C.

[Claim 19] The manufacture approach of the nature porous body of silicon carbide according to claim 17 or 18 using an alkaline-earth-metal element as this metallic element.

[Claim 20] The manufacture approach of the nature porous body of silicon carbide according to claim 19 using calcium and/or strontium as this alkaline earth metal element.

[Claim 21] The manufacture approach of the nature porous body of silicon carbide given in any 1 term of claims 17-20 chosen from the group which consists of a fluoride, carbide, a chloride, silicide, a carbonate, a hydroxide, an oxide, an inorganic-acid salt, and an organic-acid salt as a compound containing this metallic element using a kind at least.

[Claim 22] The manufacture approach of the nature porous body of silicon carbide according to claim 21 using silicate as this inorganic-acid salt.

[Claim 23] The manufacture approach of the nature porous body of silicon carbide given in any 1 term of claims 15-22 which fabricate this plastic matter in a honeycomb configuration.

[Claim 24] The manufacture approach of the nature porous body of silicon carbide given in any 1 term of claims 15-23 which carry out actual baking in a 1300-1600-degree C temperature requirement.

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**DETAILED DESCRIPTION**

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[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the nature porous body of silicon carbide used for a filter, catalyst support, etc. for motor exhaust purification, and its manufacture approach.

[0002]

[Description of the Prior Art] The porous honeycomb structure object is widely used as catalyst support for supporting the catalyst component which purifies the filter for carrying out uptake removal of the particulate matter contained in dust-containing fluid like diesel-power-plant exhaust gas, or the harmful matter in exhaust gas. Moreover, using a fireproof particle like a silicon carbide (SiC) particle as a component of such a honeycomb structure object is known.

[0003] As a concrete related technique, the silicon carbide powder which has a predetermined specific surface area and a predetermined impurity content is used as a start raw material, and the nature catalyst support of porosity silicon carbide of the honeycomb structure calcinated and acquired by the configuration of a request of this after shaping and desiccation in a 1600-2200-degree C temperature requirement is indicated by JP,6-182228,A.

[0004] Moreover, in JP,10-310474,A, calcium or calcium compound of the specified quantity is contained, and the SiC-Si compound ceramic material which shows endurance to an elevated-temperature alkali gas ambient atmosphere is indicated.

[0005]

[Problem(s) to be Solved by the Invention] Although a silicon carbide component evaporates from a silicon carbide particle front face, the neck section grows because this condenses in the contact section between particles (neck section), and an integrated state is obtained with the sintering gestalt (necking) by the recrystallization reaction of the silicon carbide powder itself shown in said JP,6-182228,A. In order for this to have to cause cost quantity since a very high burning temperature is required, and to evaporate silicon carbide and to have to carry out elevated-temperature baking of the ingredient with a high coefficient of thermal expansion, there was a problem that the baking yield fell.

[0006] Moreover, even if in charge of manufacturing the SiC-Si compound ceramic material shown in JP,10-310474,A, it is necessary to calcinate at the elevated temperature of 1800-2000 degrees C.

Furthermore, since it is the precise ingredient mainly used as baking containers, such as a crucible, this SiC-Si compound ceramic material is not employable as an ingredient for porosity filters.

[0007] this invention persons have presented the porosity honeycomb structure object containing the fireproof particle which is the aggregate especially silicon carbide, and metal silicon, and its manufacture approach in 2001 to application-for-patent 32699 specification that the above-mentioned trouble should be canceled. In the application concerned, while being able to manufacture cheaply with a comparatively low burning temperature, thermal conductivity is high and the honeycomb structure object which are fully porosity and high specific surface area is shown.

[0008] However, even if it is the manufacture approach shown in 2001 to application-for-patent 32699 specification, producing a problem depending on the case is also assumed. Drawing 4 is a mimetic diagram explaining the contact condition of the silicon carbide particle and metal silicon in the fine structure of the conventional nature porous body of silicon carbide, and metal silicon 2 contacts on the silicon carbide particle 1, and it shows the condition that both front face was covered with the oxide film 3. In addition, it is SiO<sub>2</sub> grade in the oxide film 3 here. If it calcinates according to said manufacture approach, in the nature porous body of silicon carbide from which the wettability of the metal silicon 2 and the silicon carbide particle 1 which were fused is obtained rather than may be good, the contact angle theta of the oxide film 3

on the silicon carbide particle 1 and the oxide film 3 on metal silicon 2 is an obtuse angle. That is, it has organization by which metal silicon 2 was crawled to the silicon carbide particle 1, and both touch area may be small.

[0009] Thus, when the above-mentioned touch area is small, it is assumed that the problem that the reinforcement of the nature porous body of silicon carbide itself falls, and thermal conductivity will also fall since the pass of heat conduction is also thin arises.

[0010] On the other hand, it depends for the pore diameter of the nature porous body of silicon carbide manufactured on the particle diameter of the silicon carbide particle which is the aggregate. That is, in order to obtain the nature porous body of silicon carbide with a large pore diameter, it is common to use the silicon carbide particle which has bigger particle diameter. However, shaping may become difficult if a silicon carbide particle with large particle diameter is used. Moreover, since wear of the mouthpiece for shaping also becomes remarkable when carrying out extrusion molding to a honeycomb configuration etc., there is also a possibility that problems, such as an increment in a manufacturing cost, may arise. That is, by choosing the particle diameter of a silicon carbide particle, the porosity of the nature porous body of silicon carbide is controlled, and there is a technical and manufacturing-cost-limitation in the approach of major-diameter-izing a pore diameter.

[0011] The place which this invention is made in view of the trouble which such a conventional technique has, and is made into the purpose is to offer the nature porous body of silicon carbide by which it is high porosity and high temperature conductivity while being able to manufacture cheaply, and the improvement in on the strength was made, and its manufacture approach.

[0012]

[Means for Solving the Problem] That is, according to this invention, it is a nature porous body of silicon carbide containing the silicon carbide particle used as the aggregate, and metal silicon, and the nature porous body of silicon carbide to which the average pore diameter of this nature porous body of silicon carbide is characterized by being 0.25 or more times of the mean particle diameter of this silicon carbide particle is offered.

[0013] On the other hand, according to this invention, the nature porous body of silicon carbide characterized by being a nature porous body of silicon carbide containing the silicon carbide particle used as the aggregate and metal silicon, and the contact angle of this silicon carbide particle and this metal silicon being an acute angle is offered.

[0014] Moreover, it is the nature porous body of silicon carbide which contains the silicon carbide particle used as the aggregate, and metal silicon according to this invention, and to this metal silicon of 1, when the secondary structure particle of a large number formed when these four or more silicon carbide particles contacted joins mutually together, the nature porous body of silicon carbide characterized by forming the vesicular structure is offered. In addition, in this invention, it is desirable that the number of the silicon carbide particle contained in the secondary structure particle of 1 is 30% or more of the number of the silicon carbide particle contained in the nature porous body of silicon carbide.

[0015] Furthermore, according to this invention, it is a nature porous body of silicon carbide containing the silicon carbide particle used as the aggregate, and metal silicon, and the nature porous body of silicon carbide characterized by the interfacial area of this silicon carbide particle and this metal silicon being 50% or more which doubled this interfacial area and the surface area of this metal silicon of area is offered.

[0016] In this invention, it is desirable to have the silicate compound phase of an amorphous substance or a crystalline substance on the outskirts of a front face or the outskirts of a silicon carbide particle and/or metal silicon, and it is desirable that silicon carbide particles have joined together with metal silicon and/or a silicate compound phase. Moreover, as for a silicate compound phase, it is desirable that the eutectic point of the metallic element more than a kind and a silicon dioxide is 1200-1600 degrees C including the metallic element and silicon dioxide more than kinds other than silicon, and it is still more desirable that the content to the sum total of the silicon carbide particle and metal silicon of the metallic element more than a kind is 0.1 to 10 mass %.

[0017] Furthermore, in this invention, it is desirable that a kind is an alkaline-earth-metal element at least among the metallic elements more than a kind, and it is desirable as a metallic element more than a kind to contain further metallic elements other than an alkaline-earth-metal element. Moreover, it is desirable that alkaline earth metal elements are calcium and/or strontium. In addition, in this invention, it is desirable that the content to the sum total of the silicon carbide particle and metal silicon of metal silicon is five to 50 mass %.

[0018] Moreover, according to this invention, the honeycomb structure object characterized by being



constituted by the nature porous body of silicon carbide of one of the above is offered.

[0019] On the other hand, after according to this invention adding metal silicon and an organic binder in a silicon carbide particle raw material, fabricating the plastic matter mixed, and kneaded and obtained in a predetermined configuration, carrying out temporary quenching of the acquired Plastic solid and removing the organic binder in this Plastic solid, the manufacture approach of the nature porous body of silicon carbide characterized by carrying out actual baking under a reduced pressure ambient atmosphere is offered.

[0020] Moreover, after according to this invention adding metal silicon and an organic binder in a silicon carbide particle raw material, fabricating the plastic matter mixed, and kneaded and obtained in a predetermined configuration, carrying out temporary quenching of the acquired Plastic solid and removing the organic binder in this Plastic solid, the manufacture approach of the nature porous body of silicon carbide characterized by carrying out actual baking under the reducing atmosphere containing hydrogen is offered.

[0021] After according to this invention adding the compound which contains a metallic element or this metallic element in a silicon carbide particle raw material, metal silicon, and an organic binder, fabricating the plastic matter mixed, and kneaded and obtained in a predetermined configuration, carrying out temporary quenching of the acquired Plastic solid and removing the organic binder in this Plastic solid, the manufacture approach of the nature porous body of silicon carbide characterized by carrying out actual baking under a non-oxidizing atmosphere is offered.

[0022] In this invention, it is desirable to use the metallic element whose eutectic point with a silicon dioxide is 1200-1600 degrees C, and it is desirable to use an alkaline-earth-metal element as a metallic element. Furthermore, it is desirable to use calcium and/or strontium as an alkaline earth metal element, and the thing which is chosen from the group which consists of a fluoride, carbide, a chloride, silicide, a carbonate, a hydroxide, an oxide, an inorganic-acid salt, and an organic-acid salt as a compound containing a metallic element and which use a kind at least is desirable. In addition, it is desirable to use silicate as an inorganic-acid salt.

[0023] In this invention, it is desirable to fabricate a plastic matter in a honeycomb configuration, and it is desirable to carry out actual baking in the temperature requirement which is 1300-1600 degrees C.

[0024]

[Embodiment of the Invention] Hereafter, although the gestalt of operation of this invention is explained, this invention is the range which is not limited to the gestalt of the following operations and does not deviate from the meaning of this invention, and it should be suitably understood based on this contractor's usual knowledge that modification of a design, amelioration, etc. are added.

[0025] Since the silicon carbide particle used as the aggregate and metal silicon are included, the nature porous body of silicon carbide of this invention can be sintered with a comparatively low burning temperature at the time of the manufacture, and it can raise the yield while it holds down a manufacturing cost. Moreover, even if it burns the particulate deposited for filter playback when it is used, for example for DPF (diesel particulate filter) since it has the high heat conductivity by having used metal silicon for association of the silicon carbide particle which is a fireproof particle, a local temperature rise which damages a filter does not arise.

[0026] Moreover, the nature porous body of silicon carbide of this invention requires that the average pore diameter (it is only hereafter described as a "pore diameter".) should be 0.25 or more times of the mean particle diameter (it is only hereafter described as "particle diameter".) of the silicon carbide particle which is one of the components of the nature porous body of silicon carbide concerned, it is desirable that they are 0.40 or more times, and it is still more desirable that they are 0.50 or more times. By specifying the magnitude of a pore diameter to the numeric value concerned, it is possible to consider as the nature porous body of silicon carbide which has properties, such as high porosity and high temperature conductivity, more.

[0027] In addition, since the particle size of the silicon carbide particle to be used will become large if it is going to obtain a to some extent big pore diameter when a pore diameter is less than 0.25 times of the particle diameter of a silicon carbide particle, a moldability may become poor. For example, since wear of the mouthpiece used in case extrusion molding is carried out to a honeycomb configuration etc. may become remarkable, it is not desirable.

[0028] Here, in this invention, especially the upper limit of said numeric value is not limited, and if it is the pore diameter of the same size, the particle diameter of a silicon carbide particle is so desirable that it is small. However, if substantial manufacture conditions etc. are taken into consideration, what is necessary is

just less than 5.0 times in general. in addition, the above-mentioned -- about the manufacture approach of the nature porous body of silicon carbide concerning this invention which has the structural description [ like ], it mentions later.

[0029] The nature porous body of silicon carbide of this invention is characterized by the contact angle of a silicon carbide particle and metal silicon being an acute angle. Drawing 1 is a mimetic diagram explaining an example of the contact condition of the silicon carbide particle and metal silicon in the fine structure of the nature porous body of silicon carbide of this invention, and shows the condition that metal silicon 2 contacted on the silicon carbide particle 1. That is, in the nature porous body of silicon carbide of this invention, the contact angle  $\theta$  of the silicon carbide particle 1 and metal silicon 2 is an acute angle, and the wettability of the metal silicon 2 and the silicon carbide particle 1 which were fused is improved. Therefore, since both touch area is large, the nature porous body of silicon carbide of this invention has properties, such as high intensity and high temperature conductivity, more. In addition, about the manufacture approach of the nature porous body of silicon carbide concerning this invention which has such a structural description, it mentions later.

[0030] Although it is the word used in order that a "contact angle" may usually show the include angle formed of contact into a solid-state and a liquid here, when both are solid-states, it shall use in this invention. That is, the thing of the include angle  $\theta$  which contains metal silicon among the tangent drawn to metal silicon in the point of contact of a silicon carbide particle and metal silicon and the include angle which the front face of a silicon carbide particle makes shall be called "contact angle." In addition, the thing of the include angle  $\theta$  which contains metal silicon among the tangent drawn to metal silicon and the include angle which oxide film 3 front face on the silicon carbide particle 1 makes in the point of contact of the oxide film 3 on the silicon carbide particle 1 and the oxide film 3 on metal silicon 2 in the case of the fine structure of the conventional nature porous body of silicon carbide as shown in drawing 4 shall be called "contact angle."

[0031] The nature porous body of silicon carbide of this invention is characterized by forming the vesicular structure, when the secondary structure particle (it is hereafter described as a "domain".) of a large number formed when four or more silicon carbide particles contacted joins mutually together to the metal silicon of 1. That is, in order that domains may connect mutually and they may form a vesicular structure, the amount of bond part becomes thick, and it has properties, such as high intensity and high temperature conductivity.

[0032] Moreover, when it is used, for example for DPF, it is hard to produce a local temperature rise, and it excels also in thermal shock resistance. Moreover, since pore is formed of the gap of domains, it faces pore being major-diameter-ized as compared with the case where the contact angle of a silicon carbide particle and metal silicon is an obtuse angle, and using it as a filter, and pressure loss can be made low.

[0033] Furthermore, it depends for a pore diameter on the magnitude of a domain in this invention. That is, even if it uses the silicon carbide particle which does not need to control a pore diameter by the particle diameter of the silicon carbide particle to be used, and has comparatively small particle diameter, it is possible to major-diameter-ize pore. Therefore, even if it is the case where extrusion molding is carried out to a honeycomb configuration etc., since faults, such as wear of a mouthpiece, are also controlled, improvement in the manufacture yield and reduction of facility cost are possible [ a moldability is good, and ]. in addition, the above-mentioned -- about the manufacture approach of the nature porous body of silicon carbide concerning this invention which has the structural description [ like ], it mentions later.

[0034] Furthermore, in this invention, it is desirable that the number of the silicon carbide particle contained in the secondary structure particle of 1 is 30% or more of the number of the silicon carbide particle contained in the nature porous body of silicon carbide. When it is less than 30%, the expansion effectiveness of the touch area of a silicon carbide particle and metal silicon is inadequate, and since the remarkable improvement in reinforcement, thermal conductivity, etc. is not accepted, it is not desirable.

[0035] In addition, in order to give properties, such as high porosity and high temperature conductivity, more, it is still more desirable that it is 35% or more, and it is desirable that it is especially 40% or more. Moreover, as for especially the upper limit of said numeric value, in this invention, it is desirable that it is not limited and all silicon carbide particles are contained in a secondary structure particle. However, if substantial manufacture conditions etc. are taken into consideration, what is necessary is just less than 90% in general.

[0036] On the other hand, according to this invention, it is desirable that the interfacial area of a silicon carbide particle and metal silicon needs to be 50% or more which doubled an interfacial area and the surface area of metal silicon of area, and it is 65% or more, and it is still more desirable that it is 80% or more. By specifying an interfacial area in numerical proportion concerned, it is possible to consider as the nature



porous body of silicon carbide which has properties, such as high porosity and high temperature conductivity, more. In addition, when said numeric value is less than 50%, the expansion effectiveness of the touch area of a silicon carbide particle and metal silicon is inadequate, and since the remarkable improvement in reinforcement, thermal conductivity, etc. is not accepted, it is not desirable.

[0037] Here, in this invention, especially the upper limit of said numeric value is not limited, and it is desirable that the interfacial area of a silicon carbide particle and metal silicon is 95% or more which doubled an interfacial area and the surface area of metal silicon of area. However, if substantial manufacture conditions etc. are taken into consideration, what is necessary is just less than 90% in general. In addition, about the manufacture approach of the nature porous body of silicon carbide concerning this invention which has the above structural descriptions, it mentions later.

[0038] In addition, the rate (only henceforth "the rate of an interfacial area") of the interfacial area of the silicon carbide particle and metal silicon to the area which doubled the interfacial area of the silicon carbide particle and metal silicon as used in the field of this invention and the surface area of metal silicon is computed by the following approaches. First, the target nature porous body of silicon carbide is embedded to resin, is ground, and an internal cross section is obtained. This is observed with a scanning electron microscope and image analysis of the observation photograph is performed. The interface curve of a silicon carbide particle and metal silicon and the length of a curve of a metal silicon front face are measured based on the acquired analysis photograph, and let the rate of the interface length of a curve to the sum of the interface length of a curve and the length of a curve of a metal silicon front face be the rate of an interfacial area.

[0039] In this invention, it is desirable to have the silicate compound phase of an amorphous substance or a crystalline substance on the outskirts of a front face or the outskirts of a silicon carbide particle and/or metal silicon. Drawing 2 is a mimetic diagram explaining another example of the contact condition of the silicon carbide particle and metal silicon in the fine structure of the nature porous body of silicon carbide of this invention, and shows the condition that the front face of the silicon carbide particle 1 and metal silicon 2 was covered with the silicate compound phase 4. According to an operation of the added metallic element, the oxide film on silicon carbide and the front face of a metallic element generates the silicate compound phase 4 which does not have a bad influence on wettability. Consequently, in order that the silicate compound phase 4 may show the effectiveness of improving the wettability of the silicon carbide particle 1 and metal silicon 2, both touch area is expanded. In addition, the thermal conductivity of the nature porous body of silicon carbide falls as the content of the silicate compound phase 4 increases. That is, the nature porous body of silicon carbide of this invention can show desired thermal conductivity with the content of a silicate compound phase, and this content can be adjusted suitably. Therefore, while the nature porous bodies of silicon carbide of this invention are high porosity and high intensity more, thermal properties including the thermal conductivity may be adjusted to arbitration.

[0040] In this invention, it is desirable that silicon carbide particles have joined together with metal silicon and/or a silicate compound phase. Drawing 9 is a mimetic diagram explaining the fine structure of the nature porous body of silicon carbide of this invention, silicon carbide particle 1 join mutually together with metal silicon 2 or the silicate compound phase 4, and they show the condition of having formed pore 10. Namely, the condition that silicon carbide particle 1 have combined the nature porous body of silicon carbide of this operation gestalt with metal silicon 2 ( drawing 9 (a)), The condition which silicon carbide particle 1 have combined with metal silicon 2 or metal silicon 2, and the silicate compound phase 4 ( drawing 9 (b)), Silicon carbide particle 1 may be in which condition in the condition ( drawing 9 (c)) of having joined together with metal silicon 2, the silicon carbide particle 1, the silicate compound phase 4, and the silicate compound phase 4. While the nature porous bodies of silicon carbide of this invention are high porosity and high intensity by considering as these fine structures [ like ], thermal properties including the thermal conductivity may be adjusted to arbitration. In addition, the fine structure shown in drawing 9 (a) - (c) is instantiation, and the part in the condition that this invention is not limited to these instantiation, and silicon carbide particles did not combine it, for example, metal silicon and/or a silicate compound phase adhered to the independent silicon carbide particle may be included partially.

[0041] Furthermore, in this invention, in order that that the eutectic point of the metallic element more than a kind and a silicon dioxide is 1200-1600 degrees C may aim at a more effective wettability improvement including the metallic element and silicon dioxide more than kinds other than a silicate compound phase and silicon, it is desirable. When it is less than 1200 degrees C, since the melt of MO-(M1O-, M2O-, --) SiO2 system (M, M1, M2, and -- show the metallic element more than a kind.) does not remain near the burning temperature which metal silicon fuses but it is hard to discover the wettability above-mentioned



improvement and the touch-area expansion effectiveness at the time of baking, it is not desirable. On the other hand, since the reaction of MO (M1O, M2O, --) and SiO<sub>2</sub> does not fully progress to a 1600-degree-C super-\*\*\*\*\* case at the time of baking but it is hard to discover the wettability above-mentioned improvement and the touch-area expansion effectiveness to it, it is not desirable.

[0042] Moreover, in this invention, it is desirable that the content to the sum total of the silicon carbide particle and metal silicon of the metallic element more than the above-mentioned kind is 0.1 to 10 mass %, it is still more desirable that it is 0.2 to 7 mass %, and it is desirable that it is especially 0.3 to 5 mass %. By making content of the metallic element more than a kind into said numerical range will show the effectiveness that the wettability of a silicon carbide particle and metal silicon is improved. in addition, for a certain reason, it is not desirable also when the effectiveness of a metallic element is not demonstrated, and the amount of the silicate compound phase to generate increases in a 10 mass % super-\*\*\*\*\* case too much, and it is [ come out and ] in it, in being under 0.1 mass %, and coefficient of thermal expansion increases remarkably.

[0043] In this invention, it is desirable that a kind is an alkaline-earth-metal element at least among the metallic elements more than said kind, and it is desirable as a metallic element more than a kind to contain further metallic elements other than an alkaline-earth-metal element. the wettability of a silicon carbide particle and metal silicon is effectively improved by this, and it is high porosity more -- etc. -- it is possible to consider as the nature porous body of silicon carbide which has a property.

[0044] In addition, as metallic elements other than an alkaline-earth-metal element, while forming alkaline earth metal and the system of three or more components, aluminum, Ti, Fe, etc. can be mentioned as an example that what is necessary is just that from which the eutectic point of this system becomes 1200-1600 degrees C. Furthermore, the Mg-aluminum-Si, Sr-aluminum-Si, Ba-aluminum-Si, calcium-aluminum-Si, or calcium-Mg-Si system etc. can be mentioned as an example of the system of three or more components. moreover -- cheap -- acquisition -- while it is easy, from viewpoints, such as a wettability improvement, it is desirable that said alkaline earth metal elements are calcium and/or strontium.

[0045] Furthermore, in this invention, it is desirable that the content to the sum total of the silicon carbide particle and metal silicon of metal silicon is five to 50 mass %. By making content of metal silicon into said numeric-value within the limits, as compared with the old nature porous body of silicon carbide which does not contain metal silicon, it can calcinate at low temperature and properties, such as high porosity and high temperature conductivity, are given. In addition, when the property as a binding material of metal silicon is not fully demonstrated, but it becomes inadequate joining it together according to the metal silicon of adjoining silicon carbide particles, when it is under 5 mass %, and thermal conductivity not only falls, but the structure of a thin wall like honeycomb structure is produced, since it becomes difficult to give the reinforcement which can maintain this structure, it is not desirable. Moreover, for a certain reason, it is not desirable, also when it originates in metal silicon existing, the nature porous body of silicon carbide obtained contracts too much by sintering and evils, such as a porosity fall and pore diameter contraction, occur, more than it can combine silicon carbide particles with a 50 mass % super-\*\*\*\*\* case appropriately.

[0046] On the other hand, the honeycomb structure object concerning this invention is characterized by consisting of nature porous bodies of silicon carbide of this invention mentioned above. The honeycomb structure object concerned has the outstanding oxidation resistance, acid resistance, particulate-proof reactivity, and thermal shock resistance reflecting the property of the nature porous body of silicon carbide which is the component. Furthermore, since the honeycomb structure object of this invention is a vesicular structure, it can use the particulate discharged from a diesel power plant under high SV conditions as DPF for carrying out uptake removal, catalyst support, etc.

[0047] Next, the manufacture approach of the nature porous body of silicon carbide of this invention is explained. In manufacturing the nature porous body of silicon carbide of this invention, first, metal silicon and an organic binder are added in a silicon carbide particle raw material, it mixes in it, and preparation powder is obtained. Or in using a configuration as a honeycomb structure object, metal silicon and an organic binder are added in a silicon carbide particle raw material, and it mixes and kneads in it, and obtains the plastic matter for shaping. In addition, although there is a case containing the impurity of minute amounts, such as Fe and aluminum, in the raw material used for silicon carbide particle metallurgy group silicon, you may use it as it is and what performed and refined chemical processing of chemical washing etc. may be used. Moreover, when using a honeycomb structure object as a filter, an ostomy agent may be added at the time of preparation of a plastic matter in order to raise porosity.

[0048] After fabricating said preparation powder or a plastic matter including predetermined configurations, such as a honeycomb configuration, carrying out temporary quenching of the acquired Plastic solid and

removing the organic binder in a Plastic solid (cleaning), the nature porous body of silicon carbide which has a predetermined configuration is manufactured by performing this baking under the reduced pressure ambient atmosphere of inert gas. That is, in order to calcinate under a reduced pressure ambient atmosphere according to this invention, as shown in drawing 1, the nature porous body of silicon carbide from which the oxide film of the SiO<sub>2</sub> grade shown in the front face of silicon carbide particle 1 metallurgy group silicon 2 was volatilized and removed can be obtained.

[0049] Since an oxide film becomes the cause that the wettability of a silicon carbide particle and metal silicon falls, it is possible by volatilizing and removing this to make the touch area of a silicon carbide particle and metal silicon increase. Therefore, the nature porous body of silicon carbide reinforcement and whose thermal conductivity improved can be manufactured.

[0050] In addition, the above-mentioned reduced pressure ambient atmosphere should just be a reduced pressure ambient atmosphere whose oxide film of SiO<sub>2</sub> grade is extent which can be volatilized and removed by calcinating. 0.1-10hPa is desirable and, specifically, 0.5-5hPa is still more desirable. It is difficult in facility cost to be referred to as less than 0.1hPa, and if it 10hPa super-\*\*, since it becomes inadequate an oxide film's volatilizing, it is not desirable. Moreover, although not limited especially about the inert gas kind for considering as a non-oxidizing atmosphere, either, it is desirable to use Ar from viewpoints, such as acquisition and handling ease.

[0051] Next, another operation gestalt of the manufacture approach of the nature porous body of silicon carbide concerning this invention is explained. That is, after fabricating said preparation powder or a plastic matter including predetermined configurations, such as a honeycomb configuration, carrying out temporary quenching of the acquired Plastic solid and removing the organic binder in a Plastic solid (cleaning), the nature porous body of silicon carbide which has a predetermined configuration is manufactured by carrying out actual baking under the reducing atmosphere containing hydrogen.

[0052] Drawing 3 is a mimetic diagram explaining still more nearly another example of the contact condition of the silicon carbide particle and metal silicon in the fine structure of the nature porous body of silicon carbide of this invention, returns the oxide film of the SiO<sub>2</sub> grade shown in the front face of silicon carbide particle 1 metallurgy group silicon 2 to removal, returns SiO<sub>2</sub> to Si in reduction, and is taken as new metal silicon (returned metal silicon 5). Therefore, since the wettability of the silicon carbide particle 1 and metal silicon 2 is improved, the touch area of the silicon carbide particle 1 and metal silicon 2 increases, and the nature porous body of silicon carbide reinforcement and whose thermal conductivity improved can be manufactured.

[0053] In addition, in this invention, although especially the hydrogen content at the time of baking is not limited, the oxide film of SiO<sub>2</sub> grade should just be the hydrogen content of extent removable in reduction by calcinating. 0.5 - 10 volume % is desirable, and, specifically, 1 - 4 volume % is still more desirable. Under 0.5 volume % of reduction-removal of an oxide film is inadequate, and since the handling of hydrogen gas becomes difficult, 10 volume % super-\*\*(ing) is not desirable.

[0054] Moreover, still more nearly another operation gestalt of the manufacture approach of the nature porous body of silicon carbide concerning this invention is explained. That is, in a silicon carbide particle raw material, with metal silicon and an organic binder, the compound containing a metallic element or its metallic element is added, it mixes, and preparation powder is obtained. After fabricating this preparation powder or plastic matter including predetermined configurations, such as a honeycomb configuration, carrying out temporary quenching of the acquired Plastic solid and removing the organic binder in a Plastic solid (cleaning), the nature porous body of silicon carbide which has a predetermined configuration is manufactured by carrying out actual baking under a non-oxidizing atmosphere.

[0055] That is, in order to add a metallic element or the compound containing the metallic element in raw materials, such as a silicon carbide particle, according to this invention, a presentation and condition of an oxide film change. Therefore, as shown in drawing 3, since the front face of the silicon carbide particle 1 and metal silicon 2 is covered with the silicate compound phase 4, the wettability of the silicon carbide particle 1 and metal silicon 2 is improved, and it can manufacture the nature porous body of silicon carbide reinforcement and whose thermal conductivity improved.

[0056] Here, in this invention, the effectiveness at the time of adding a metallic element or the compound containing the metallic element is explained to raw materials, such as a silicon carbide particle. The oxide film in front faces, such as a silicon carbide particle, moves to each front face from the interface of a silicon carbide particle and metal silicon at the time of baking. At this time, an oxide film 2, i.e., SiO, reacts with a metallic element, it forms the compound of MO-(M1O-, M2O-, --) SiO<sub>2</sub> system (M, M1, M2, and -- show the metallic element more than a kind.), and is fixed by the crystalline substance or the vitreous state



(amorphous) as a phase which is different in SiO<sub>2</sub>. Consequently, the wettability of a silicon carbide particle and metal silicon will improve.

[0057] In addition, it is desirable that the eutectic point of SiO<sub>2</sub> is 1200-1600 degrees C, as for said metallic element, it is still more desirable that it is 1300-1550 degrees C, and it is desirable that it is especially 1350-1500 degrees C. When it is less than 1200 degrees C, since the melt of MO-(M1O-, M2O-, --) SiO<sub>2</sub> system does not remain near the burning temperature which metal silicon fuses but it is hard to discover the wettability above-mentioned improvement and the touch-area expansion effectiveness at the time of baking, it is not desirable. On the other hand, since the reaction of MO (M1O, M2O, --) and SiO<sub>2</sub> does not fully progress to a 1600-degree-C super-\*\*\*\*\* case at the time of baking but it is hard to discover the wettability above-mentioned improvement and the touch-area expansion effectiveness to it, it is not desirable.

[0058] It may not be limited especially about the gestalt of the compound containing a metallic element and a metallic element, and powder or a solution is sufficient. Furthermore, as the metallic element concerned, it is desirable to use an alkaline-earth-metal element, and especially as an alkaline-earth-metal element, it is desirable to use calcium and/or strontium in order to aim at the further wettability improvement and exertion of the touch-area expansion effectiveness. Moreover, handling and the acquisition of calcium are desirable also in an easy point.

[0059] As a compound containing a metallic element, it is independent about the fluoride of a metallic element, carbide, a chloride, silicide, a carbonate, a hydroxide, an oxide, an inorganic-acid salt, and an organic-acid salt, or such mixture can be used. Furthermore, it is desirable silicate and to specifically use cordierite (Mg-aluminum silicate) and anorthite (calcium-aluminum silicate) as the above-mentioned inorganic-acid salt. These compounds are cheap, and since they can make more the nature porous body of silicon carbide obtained high porosity and high intensity and can adjust thermal properties including the thermal conductivity to arbitration, they are desirable while acquisition is easy. In addition, while it is stable in atmospheric air, to say nothing of what a toxic gas etc. does not generate in a baking process being desirable, a carbonate, an oxide, or a hydroxide is desirable.

[0060] In addition, as for temporary quenching, in the manufacture approach of the nature porous body of silicon carbide of this invention, it is desirable to carry out at temperature lower than the temperature which metal silicon fuses. You may once hold at the predetermined temperature of about 150-700 degrees C, and to below 50 degrees C / hr, a programming rate may be made late and, specifically, may carry out temporary quenching in a predetermined temperature region. Moreover, about the technique once held at predetermined temperature, with the class and amount of an organic binder which were used, maintenance or maintenance with the two or more temperature level of only a 1 temperature level is sufficient, and in holding with the two or more temperature level further, even if the same, you may change the holding time mutually. Moreover, between a certain 1 temperature-province regions may be similarly made late about the technique of making a programming rate late, or you may make it late among the two or more division, and, in between the two or more [ further ] division, a rate may be mutually changed also as the same.

[0061] Metal silicon needs to become soft in order to obtain the organization where the fireproof particle was combined with metal silicon. It is desirable to carry out actual baking above 1410 degrees C, in order to obtain the organization combined only with metal silicon, since the melting point of metal silicon is 1410 degrees C, and in order to obtain the organization combined by the silicate compound phase, it is desirable to carry out actual baking above 1300 degrees C. Moreover, the optimal burning temperature is determined also from the microstructure and characteristic value of an organization. However, since association which evaporation of metal silicon advanced at the temperature exceeding 1600 degrees C, and minded metal silicon becomes difficult, 1300-1600 degrees C is suitable for the temperature of this baking, and it is desirable. [ of 1350-1550 degrees C ]

[0062] In addition, according to the manufacture approach using the recrystallizing method shown in aforementioned JP,6-182228,A, since it has the structure combined by silicon carbide particles, the sintered compact of high thermal conductivity is obtained, but Since it sinters by the evaporation condensation device in which it stated previously, in order to evaporate silicon carbide A burning temperature higher than the manufacture approach of this invention is needed, and in order to obtain the practically usable nature porous body of silicon carbide, it is usually necessary to calcinate at least 1800 degrees C or more at an elevated temperature 2000 degrees C or more.

[0063]

[Example] Hereafter, although this invention is further explained to a detail based on an example, this invention is not limited to these examples.

[0064] (Example 1) SiC raw material powder with a mean particle diameter of 32.6 micrometers and Si



powder with a mean particle diameter of 4 micrometers were blended so that it might become the presentation of 80:20 with a mass ratio, and the methyl cellulose 6 mass section, the surfactant 2.5 mass section, and the water 24 mass section were added as an organic binder to this powder 100 mass section, it mixed and kneaded to homogeneity, and the plastic matter for shaping was obtained. The obtained plastic matter was fabricated with the extruding press machine in 0.43mm in the outer diameter of 45mm, die length of 120mm, and septum thickness, and the honeycomb configuration of cel consistency 100 cel / square inch (16 cels / cm<sup>2</sup>).

[0065] After performing temporary quenching for cleaning for this honeycomb Plastic solid at a hypoxia ambient atmosphere and 550 degrees C for 3 hours, in 2hPa Ar ambient atmosphere, baking of 2 hours was performed at 1450 degrees C, and the nature porous body of silicon carbide of honeycomb structure was produced by porosity. Moreover, the microphotography which is the fine structure of the nature porous body of silicon carbide obtained by drawing 5 is shown. In addition, for a gray part, a silicon carbide particle and a white part are [ metal silicon and a black part ] pores among drawing. About the following and a microphotography, it is the same.

[0066] (Example 2) Temporary quenching was performed like said example 1. In Ar ambient atmosphere which included the acquired temporary-quenching object for 4% of hydrogen, baking of 2 hours was performed at 1450 degrees C, and the nature porous body of silicon carbide of honeycomb structure was produced by porosity. Moreover, the microphotography which is the fine structure of the nature porous body of silicon carbide obtained by drawing 6 is shown.

[0067] (Example 3) SiC raw material powder with a mean particle diameter of 32.6 micrometers and Si powder with a mean particle diameter of 4 micrometers were blended so that it might become the presentation of 80:20 with a mass ratio, and to this powder 100 mass section, 1 mass % (it is 0.4 mass % by calcium conversion) addition of the calcium carbonate was carried out, and it mixed. Furthermore, to this powder 100 mass section, the methyl cellulose 6 mass section, the surfactant 2.5 mass section, and the water 24 mass section were added as an organic binder, it mixed and kneaded to homogeneity, and the plastic matter for shaping was obtained. The obtained plastic matter was fabricated with the extruding press machine in 0.43mm in the outer diameter of 45mm, die length of 120mm, and septum thickness, and the honeycomb configuration of cel consistency 100 cel / square inch (16 cels / cm<sup>2</sup>).

[0068] After performing temporary quenching for cleaning for this honeycomb Plastic solid at a hypoxia ambient atmosphere and 550 degrees C for 3 hours, in Ar ambient atmosphere, baking of 2 hours was performed at 1450 degrees C, and the nature porous body of silicon carbide of honeycomb structure was produced by porosity. Moreover, the microphotography which is the fine structure of the nature porous body of silicon carbide obtained by drawing 7 is shown.

[0069] (Example 4) The calcium carbonate was replaced with doing 1 mass % (it being 0.4 mass % by calcium conversion) addition of, and the nature porous body of silicon carbide of honeycomb structure was produced by porosity by the same approach as said example 3 except doing 1 mass % (it being 0.6 mass % by strontium conversion) addition of a strontium carbonate.

[0070] (Example 5) SiC raw material powder and Si powder were replaced with blending so that it may become the presentation of 80:20 with a mass ratio, and the nature porous body of silicon carbide of honeycomb structure was produced by porosity by the same approach as said example 3 except blending SiC raw material powder, Si powder, and cordierite (Mg-aluminum silicate) powder so that it may become the presentation of 80:10:10 with a mass ratio.

[0071] (Example 1 of a comparison) Temporary quenching was performed like said example 1. In ordinary pressure and Ar ambient atmosphere, baking of 2 hours was performed for the acquired temporary-quenching object at 1450 degrees C, and the nature porous body of silicon carbide of honeycomb structure was produced by porosity. Moreover, the microphotography which is the fine structure of the nature porous body of silicon carbide obtained by drawing 8 is shown.

[0072] (Physical property trial) each nature porous body of silicon carbide produced in the above-mentioned examples 1-3 and the example 1 of a comparison -- a method of mercury penetration -- an average pore diameter -- measuring -- Archimedes -- porosity was measured by law. Moreover, the predetermined test piece was cut down, the three-point bending test using a material testing machine was performed in the bottom of a room temperature condition, and reinforcement was measured. Furthermore, thermal conductivity was measured with the laser flash method. A result is shown in Table 1.

[0073]

[Table 1]

	平均気孔径 ( $\mu\text{m}$ )	気孔率 (%)	強度 (MPa)	熱伝導率 (W/mK)
実施例 1	15	53	13	21
実施例 2	15	51	19	17
実施例 3	14	50	23	16
実施例 4	13	49	27	16
実施例 5	15	48	34	8
比較例 1	7	42	11	14

[0074] As shown in Table 1, the nature porous body of silicon carbide concerning this invention of examples 1-5 has a large average pore diameter as compared with the nature porous body of silicon carbide of the example 1 of a comparison, and is high porosity. Furthermore, the nature porous body of silicon carbide of examples 1-4 was able to check the effectiveness which excels [ thermal conductivity / reinforcement and ] in thermal shock resistance as compared with the example 1 of a comparison since it is high, and was excellent in this invention.

[0075] On the other hand, though the nature porous bodies of silicon carbide of an example 5 are an average pore diameter equivalent to the nature porous body of silicon carbide of examples 1-4, porosity, and reinforcement, it turns out that thermal conductivity is low. That is, it originates in the nature porous body of silicon carbide of an example 5 containing the cordierite which is Mg-aluminum silicate, and it is thought that the thermal conductivity fell. Therefore, it became clear that it is possible to control suitably the pore properties (an average pore diameter, porosity, etc.) of the nature porous body of silicon carbide obtained, mechanical properties (reinforcement etc.), and thermal properties (thermal conductivity etc.) according to an application by adjusting the ingredient to be used and its presentation.

[0076] Moreover, the contact angle of a silicon carbide particle and metal silicon is an acute angle, and the nature porous body of silicon carbide concerning this invention of examples 1-3 has a large touch area, as shown in drawing 5 -7. Furthermore, pore is formed of the gap of domains and it was able to check that pore was major-diameter-sized as compared with the case where the contact angle of a silicon carbide particle and metal silicon as shown in drawing 8 is an obtuse angle.

[0077]

[Effect of the Invention] As explained above, though the fireproof particle like a silicon carbide particle is included, since it can be made to sinter with a comparatively low burning temperature at the time of the manufacture, the nature porous body of silicon carbide and honeycomb structure object of this invention can improve [ yield's ] while holding down a manufacturing cost, and can be offered cheaply. Moreover, since it has the predetermined fine structure, are the nature porous body of silicon carbide which has high porosity, high temperature conductivity, and the property of high intensity, for example, when it is used for DPF, it sets. Even if it burns the particulate deposited for filter playback, since it is a porous honeycomb structure object further, by not producing local generation of heat which damages a filter It can be suitably used also under high SV conditions as a filter, catalyst support, etc. for motor exhaust purification.

[0078] Furthermore, the manufacture approach of the nature porous body of silicon carbide of this invention can manufacture cheaply the nature porous body of silicon carbide by which it is high porosity and high temperature conductivity, and the improvement in on the strength was made according to a predetermined process and predetermined conditions.

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[Translation done.]

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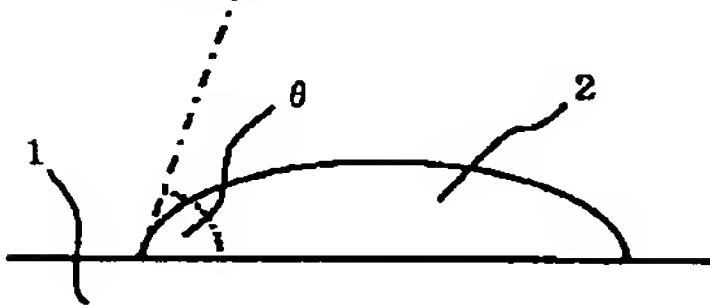
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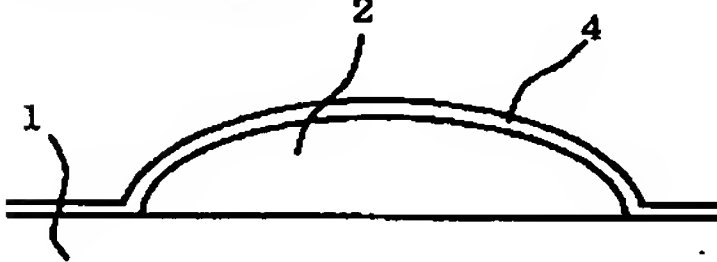
DRAWINGS

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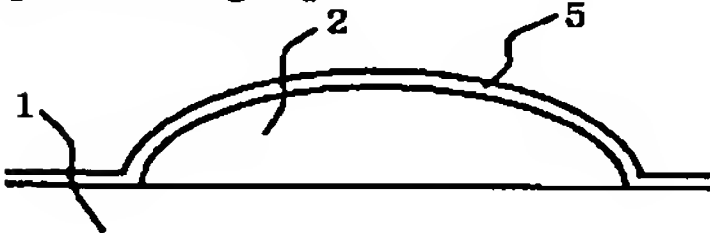
[Drawing 1]



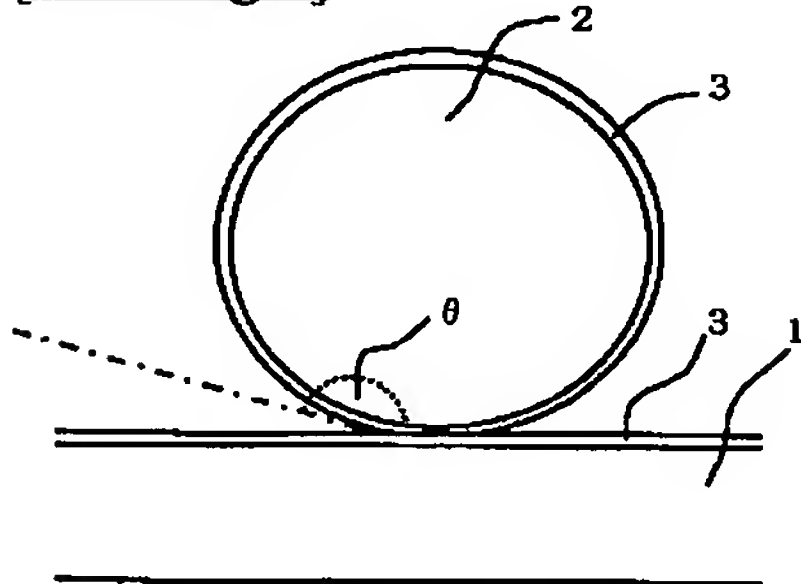
[Drawing 2]



[Drawing 3]



[Drawing 4]

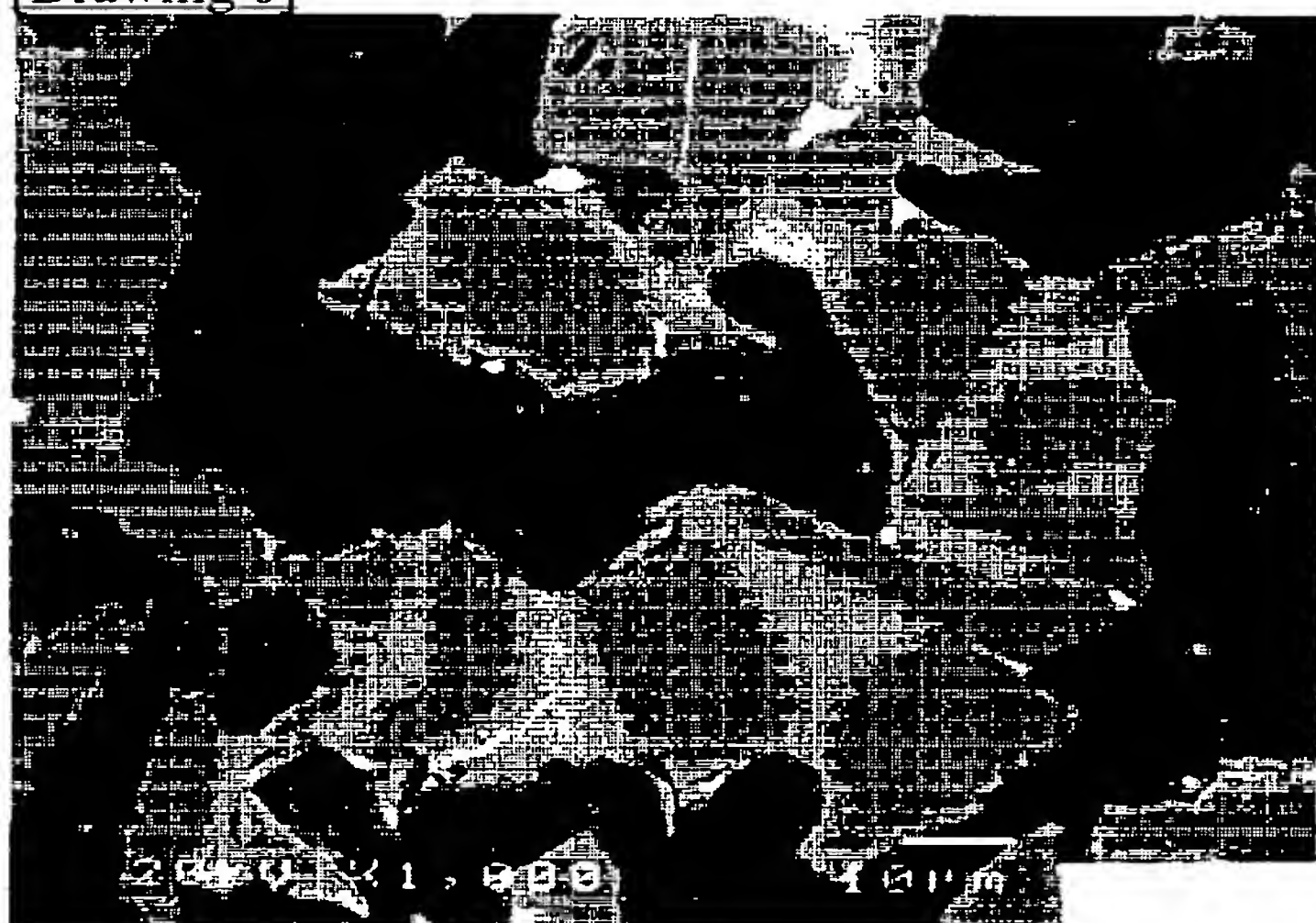


[Drawing 5]





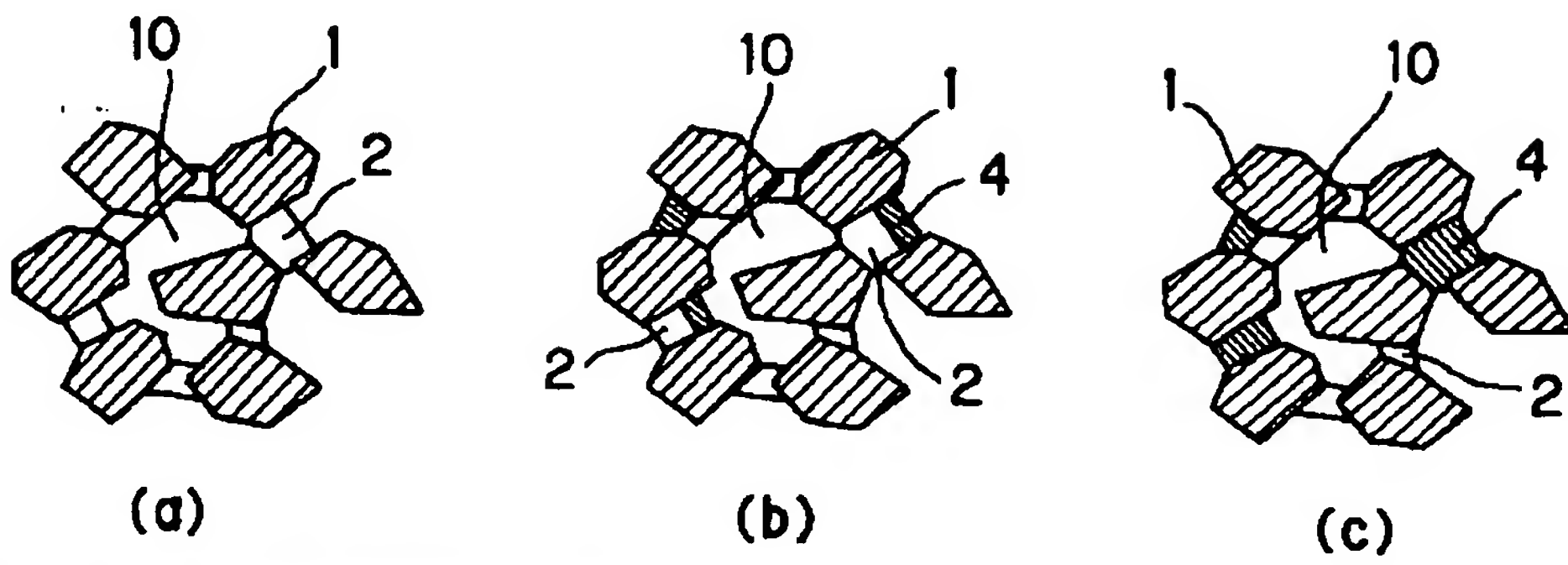
[Drawing 6]



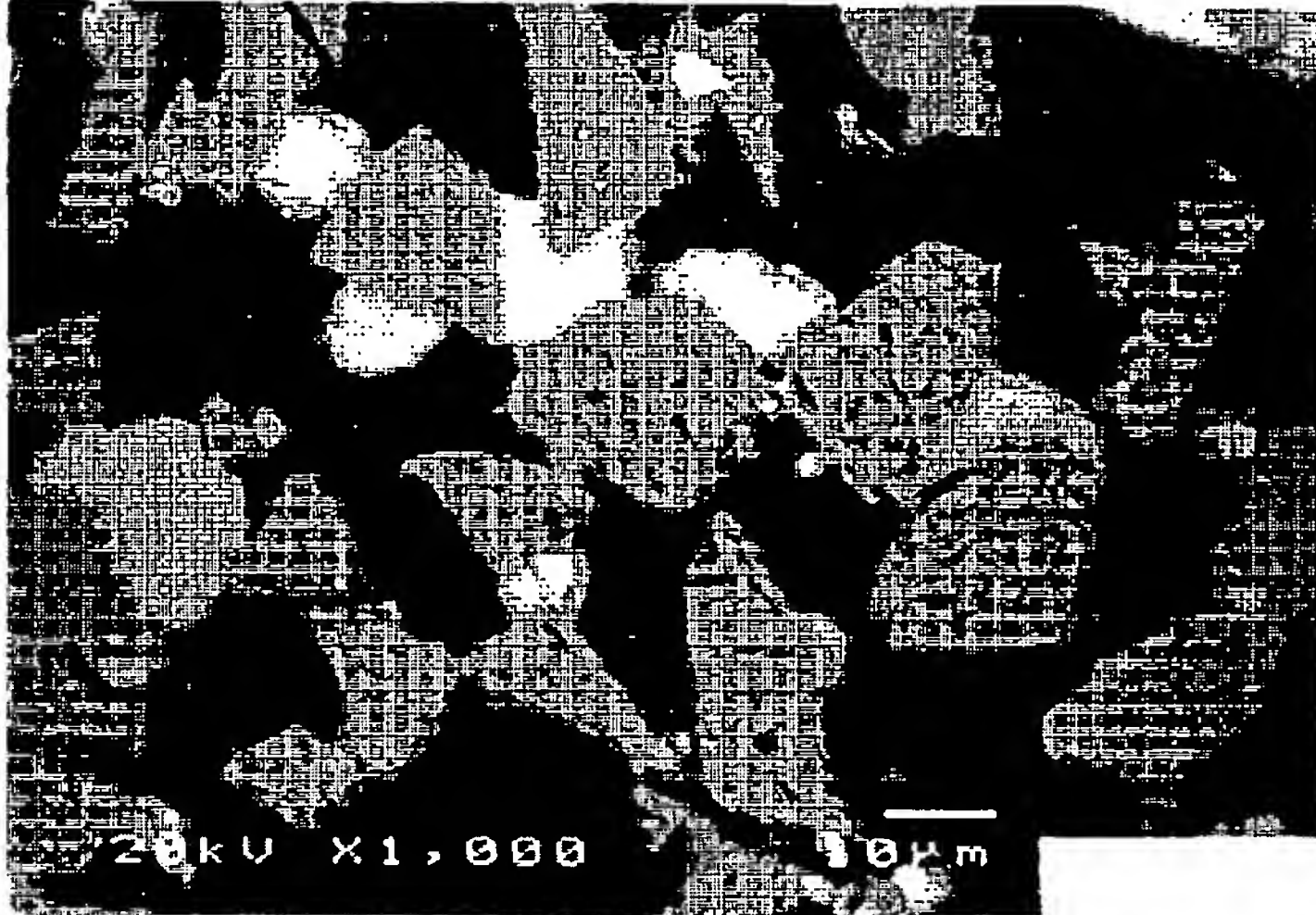
[Drawing 7]



[Drawing 9]



[Drawing 8]



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[Translation done.]